PROTOTYPE DEMONSTRATION OF GAMMA INSENSITIVE CENTRIFUGALLY TENSIONED METASTABLE FLUID NEUTRON/ALPHA DETECTOR

A Thesis

by

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ABSTRACT

Tensioned metastable fluid detectors are a novel detector concept with a wide array of potential uses. A particularly interesting application is as a real-time alpha and neutron detector that is also insensitive to gamma rays. This type of detector could have many uses for safeguards and nonproliferation including reprocessing facilities and border protection. For safeguards facilities, the detector could be used to perform online measurements of alpha-emitting nuclides such as uranium and plutonium in the reprocessing fluids. The online measurements could include dissolver tank fluids and products solution fluids where conventional detector use is infeasible.

In this research, an initial characterization of one version of the tensioned metastable fluid detector (TMFD) was performed. A TMFD works by putting a fluid under negative pressure or tension. Once under tension, small amounts of energy can cause the tensioned molecules in the fluid to burst apart forming a visible bubble. The small amount of energy can come from nuclear particles such as alphas and neutrons. Another key concept of the TMFD is that the amount of energy needed to form a bubble is dependent on the negative pressure of the fluid. By varying the negative pressure, only particles of a desired energy or higher would form bubbles in the system allowing for a form of spectroscopy. The spectroscopy could be used, along with a volume measurement, to quantify the amount of uranium or plutonium in the reprocessing fluid.

The initial characterization was performed using gamma, neutron, and alpha radiation. The gamma-ray tests were performed using an external ^{137}Cs source next to

the system. Several neutron tests were performed using an external ^{252}Cf source including a test to verify compliance with the inverse square law and a test varying the pressure of the TMFD to test spectroscopy uses. Last, alpha-radiation tests were conducted using various amounts of depleted uranium dissolved in the working fluid of the detector. This study confirmed that the detector system is insensitive to low amounts of gamma-rays. It also confirmed that by varying the negative pressure in the detector, different energy neutrons could be detected. Last, the study confirmed that the detector could be used to detect alpha particles in real time but that additional work was needed on the design to ensure reliable results.

The TMFD can be used to detect alpha particles and neutrons and is insensitive to gamma-rays. However, the current design of the system is a prototype and must be redesigned in order to be used in a reprocessing facility. Also, a lower negative pressure is needed for neutron detection compared to alpha detection. The pressure discrepancy would cause the detector to be unable to detect alpha particles in a large neutron radiation field making the detector unusable at the dissolver tank, but still usable for the reprocessing product lines.

DEDICATION

To my grandfather, Les Martin, for inspiring me to become an engineer.

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NOMENCLATURE

TMFD	Tensioned Metastable Fluid Detector
CTMFD	Centrifugally Tensioned Metastable Fluid Detector
ATMFD	Acoustically Tensioned Metastable Fluid Detector
RPS	Revolutions Per Second
WT	Wait Time
RT	Run Time
Test	A batch of runs
Experiment	A batch of tests

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1. INTRODUCTION

There are currently two major commercial reprocessing facilities for used nuclear fuel in the world: the AREVA La Hague plant in France and the Rokkasho Nuclear Fuel Reprocessing Facility in Japan. These facilities use a variation of the PUREX, Plutonium Uranium Redox Extraction, method to take used fuel from reactors and remove the uranium and plutonium for use in fresh mixed oxide fuel. Safeguards are needed at these facilities to ensure that none of the plutonium or uranium is diverted or stolen.

PUREX takes spent nuclear fuel assemblies, separates the fuel from the casing, melts the fuel using nitric acid into a dissolver tank, uses tributyl phosphate to separate the uranium and plutonium from the fission products, and then separates the uranium from the plutonium. Fig. 1 shows a diagram of PUREX along with current safeguards.[1] While in aqueous form, current safeguards consist of non-destructive analysis (NDA) methods. At the aqueous volume measurement points, after the dissolver tank and in the product feed lines, a sample of the aqueous feed is removed from the system and a Hybrid K-edge Densitometer (HKED) is used to measure the amount of uranium and plutonium in the aqueous feed.[2] The HKED uses a K-edge densitometer (KED) enhanced with x-ray fluorescence (XRF) to measure the amount of uranium in the sample and the PU/U ratio.[3] However, this measurement cannot be performed continuously as a sample has to be taken from the aqueous stream and is not returned.

A tensioned metastable fluid detector (TMFD) could be used instead of the HKED. The TMFD would be placed online meaning the TMFD would, autonomously, take fluid from the aqueous line, test the fluid for uranium and plutonium concentration,

and return the fluid to the aqueous line. The online deployment along with the already performed volume measurements could be used to determine the amount of plutonium and uranium at any time without waste. The optimal placement for the TMFD would be at the dissolver tank, which contains all of the dissolved used fuel including plutonium uranium and fission products, and in the uranium and plutonium product feed lines, which only contain uranium and plutonium, respectively.

For this research, a centrifugally tensioned metastable fluid detector (CTMFD) was procured and characterized at Texas A&M University. A series of tests were performed to determine the CTMFD's capabilities in regards to gamma-ray detection, neutron spectroscopy, and alpha quantification. The capabilities were studied to determine the usefulness of a CTMFD system for safeguards use at a reprocessing facility.

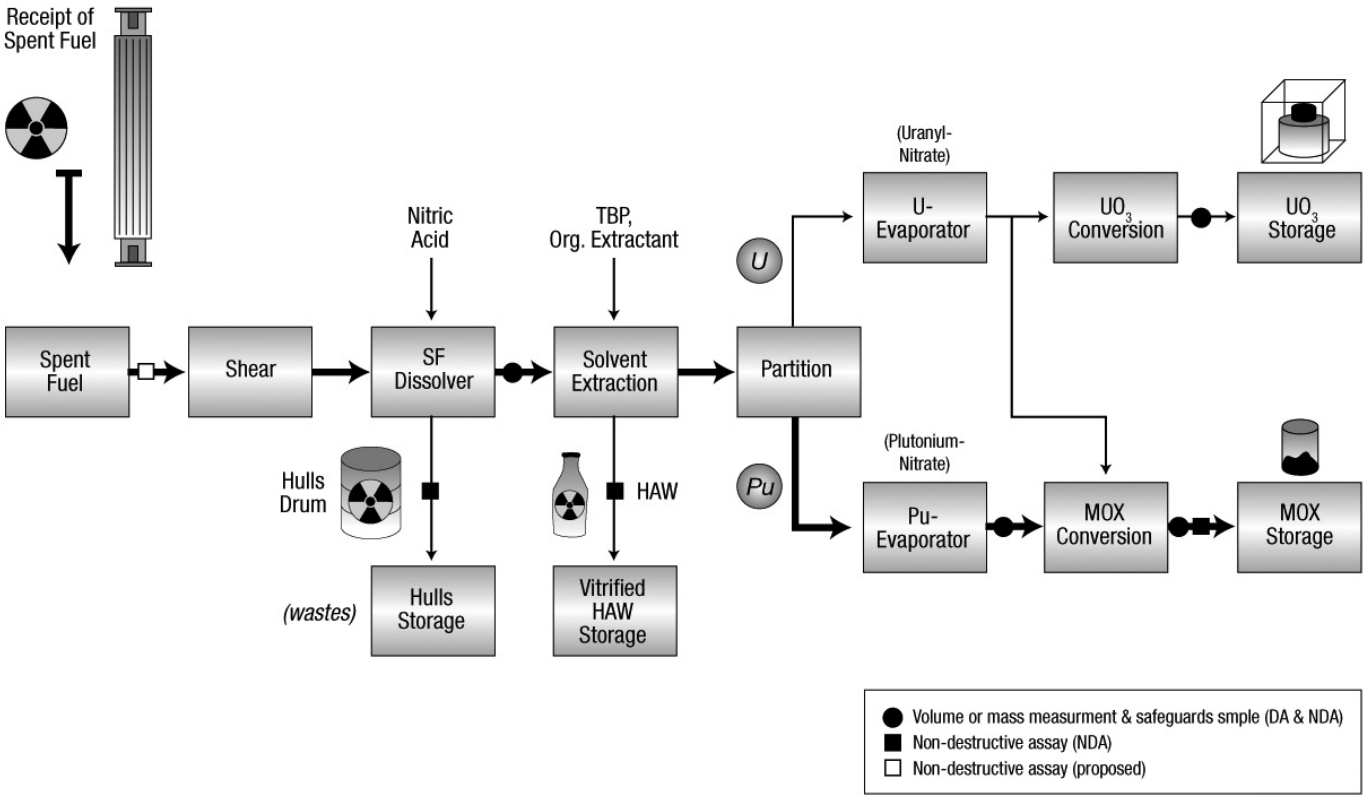

Fig. 1. Diagram of the PUREX Process [1]

2. BACKGROUND AND THEORY

The first bubble chamber was used by Dr. Glaser in 1952. In his work, Dr. Glaser placed a fluid in a metastable region by superheating the fluid. He saw that exposing superheated diethyl ether, at a pressure of twenty atmospheres, to a ^{60}Co source caused the rapid formation of bubbles once the pressure was removed.[4] In 1958, Dr. Seitz developed the thermal spike theory which describes the mechanism for the formation of bubbles in a superheated fluid. In his work, Dr. Seitz theorized that ionizing radiation causes temperature spikes in the superheated fluid which causes bubbles to form from the evaporation of the fluid.[5]

Initial research and work on bubble chambers using tensioned fluids and their applications were completed by Dr. Rusi Taleyarkhan and his students at Purdue University. They have completed the initial theory work behind the system as well as designed and built several prototype systems. The system being tested at Texas A&M University was procured from Sagamore-Adams Laboratories, LLC.

2.1 Bubble Formation and Tension Metastability

The main concept behind TMFDs is the tensioning of a working fluid. Similar to solids, liquids can be put under tension which stretches the bonds between molecules in the fluid. A semi-equilibrium state can be achieved, called metastability, where the bonds are stretched, but without additional energy the bonds will not break. Once a small amount of energy is added to this metastable system, the bonds will break forming

nanoscale vapor bubbles.[6] The process can be better described using a P-V diagram as shown in Fig. 2.

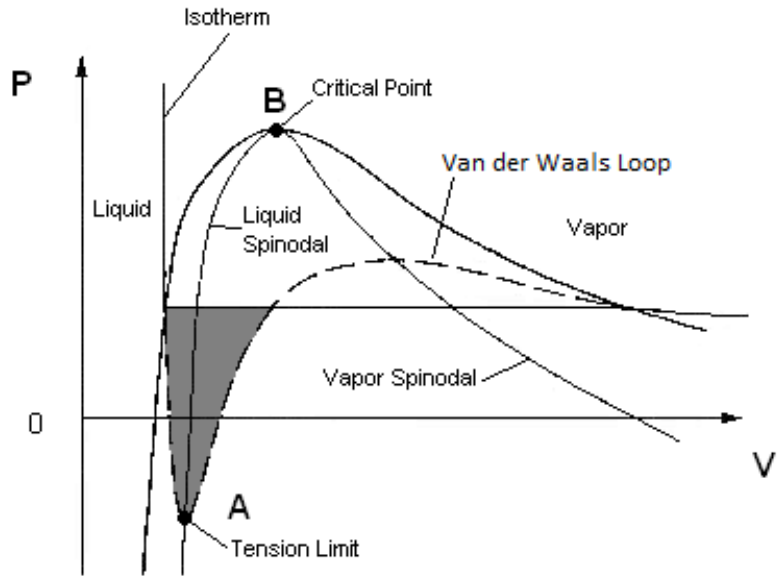

Fig. 2. Liquid-Vapor Phase Diagram Showing the Metastable Region [7]

In this figure, the grayed area shows where the fluid can be metastable. The area is inside the two phase region and is bound by the isotherm, a constant temperature, and the Van der Waals loop, beyond which, a fluid must be two-phased. Normally, inside this region, the fluid must contain vapor, but by tensioning, the fluid can be brought down into this region without vapor forming. Once in this region, the addition of small amounts of energy will cause nanoscale vapor bubbles to form. These nanoscale vapor bubbles can become macroscopic bubbles if enough energy is added for the bubbles to reach a critical radius. The amount of energy needed to form bubbles has been estimated based on past results and is confirmed with experimentation. These macroscopic bubbles

indicate the deposition of energy by a nuclear particle (neutron, alpha particle, beta particle or gamma ray) and are the basis behind the TMFD.[8]

Gamma rays interact and lose energy via three main methods photoelectric absorption, Compton scattering, and pair production. First, in photoelectric absorption, the gamma ray is fully absorbed by the atom and an energetic photoelectron is released. Next, in Compton scattering, the gamma ray reacts directly with an electron, imparting some of its energy, which ejects the electron from the atom. Last, is pair production, if the gamma ray has an energy greater than 1.022 MeV, the gamma ray can interact with the Columbic field around the nucleus and convert to an electron-positron pair.[9]

Neutrons interact with matter in one of two main ways depending on their energy. First, fast neutrons will recoil off the nucleus of an atom. The recoil imparts a large amount of energy to the recoil nucleus. Second, slow neutrons will either elastically bounce off of a nucleus imparting small amounts of energy, or the neutron will be absorbed and cause a nuclear reaction. These nuclear reactions often involve the production of energetic charged particles (in the case of B or Li) or recoil nuclei (in the case of radiative capture reactions in other nuclei).[9]

Charged particles (alphas and betas) react mainly in one way. When the charged particle passes near an atom, it reacts with the columbic force of the electrons and either excites the electron to a higher shell or it imparts enough energy to completely remove the electron from the atom. Due to their large size, alpha particles lose their energy much faster than betas and therefore can only travel a few centimeters in air.[9]

In the CTMFD, enough energy needs to be deposited within hundreds of nanometers in order for the tensioned bonds to be broken.[10] Most nuclear particles deposit their energy over several reactions spanning a distance greater than one hundred nm. However, ionized recoil nuclei, caused by particle interactions or nuclear particle formation, are massive enough that their energy can be deposited over a very short distance. This is the reason alpha particles and neutrons are detected by the TMFD but gamma-rays are not (since gamma rays almost solely interact with electrons).[11] Neutrons form recoil nuclei when interacting with the working fluid and alpha emitters are placed directly in the fluid, due to the short range of alpha particles, so the recoil nuclei from the creation of the alpha particle can deposit enough energy to form bubbles.

2.2 CTMFD System

Currently there are two methods for tensioning a fluid. The acoustically tensioned metastable fluid detector (ATMFD) uses acoustic waves to tension the fluid. Using a simpler approach, the CTMFD rapidly spins the fluid causing tension through centrifugal force. In this research, a CTMFD was used.[11] The general design of a CTMFD spinner can be seen in Fig. 3.

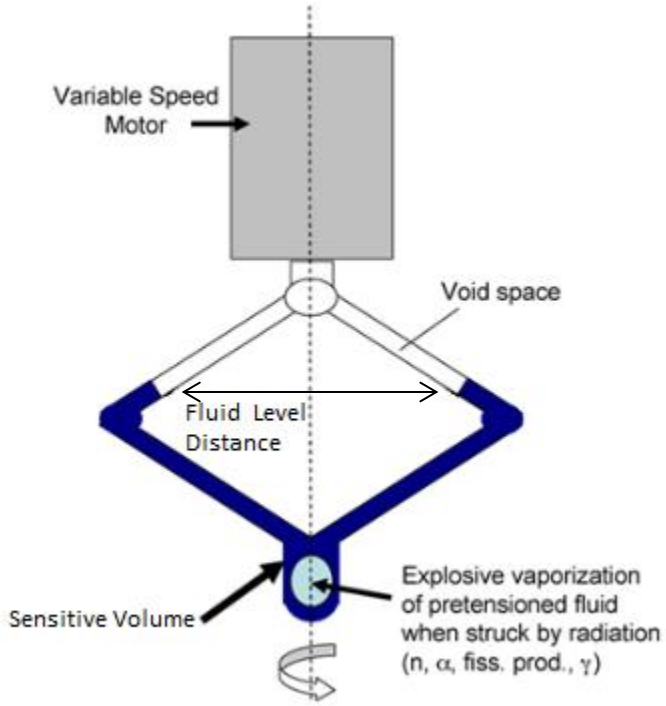

Fig. 3. Diagram of CTMFD Spinner [10]

The CTMFD uses a rotary tool to rapidly rotate the spinner about a central axis. This causes the fluid in the sensitive volume to be placed under tension. A nuclear event of the necessary energy will cause a bubble to be formed in the sensitive volume. The energy needed to cause a macroscopic bubble to form can be varied by changing the negative pressure in the sensitive volume through adjustments in the revolutions per second (RPS). Controlling the negative pressure can be used to setup the system so that only particles of a certain energy or higher will form bubbles. This technique can be used to perform limited nuclear spectroscopy.[11]

Spectroscopy using the CTMFD can be completed in a step wise manner. At each negative pressure only particles of a certain energy or above can form a bubble in

the CTMFD. As the negative pressure in the system is increased, particles of decreasing energy will cause detections in the system. Based on the time it takes a bubble to form in the system at each pressure, the activity of the material can be calculated. However, because energies greater than the required energy can still form bubbles, spectroscopy must be done in a stepwise format. First, the lowest pressure at which bubbles form is used and a count rate (C_1) is acquired. Next, the pressure is increased and a new count rate (C_2) is acquired. However, the new count rate also includes the energies from the first count, so C_1 must be subtracted in order to get the count rate due only to those reactions from particles above the energy for C_1 and below the energy for C_2. This process is repeated by increasing the pressure until either the maximum allowable pressure is reached or the count rate stops increasing suggesting that there are no particles in the radiation field above the last energy. The activity (A_i) for each energy group *i* can then be acquired, assuming a constant efficiency η for the CTMFD, via unfolding using the following series of equations:

$$A_1 = C_1/\eta \tag{1}$$

$$A_2 = (C_2 - C_1)/\eta \tag{2}$$

$$A_i = (C_i - C_{i-1})/\eta \tag{3}$$

The activities are then plotted to acquire a spectrum. It should be noted that the assumption of a constant efficiency is an untested assumption and is left as future work.

The method used to detect radiation by TMFDs is very different than that used by conventional detectors. In ion chambers, ionizing radiation passes through a gas causing an ion pair to be formed. When the ion pair is formed, the charge is collected using an electric field and signifies the detection of radiation. In scintillator detectors, ionizing radiation enters the scintillator which creates a light photon. The photon strikes a photocathode and creates an electron through the photoelectric effect. The electron is multiplied using a series of dynodes in the photomultiplier tube and the resulting electrons have a large enough charge to be detected signifying the detection of radiation. In semiconductor detectors, ionizing radiation creates free electrons and holes in the semiconductor material. The electrons and holes travel in opposite directions towards electrodes where a pulse is created that can be detected signifying the detection of radiation.[9]

3. EXPERIMENTAL PROCEDURE

Several tests were performed with the CTMFD. Initial tests were performed to acquire familiarity with the system. The tests included finding the optimal method for filling the spinner, finding the optimal method for attaching the reflective tape, experimenting to determine how the data were presented after a test, determining an optimal testing method, acquiring a general familiarity with preparing the tests, and use of the software. Following the initial tests, formal tests were completed using a strict method to ensure the results were comparable and repeatable.

First, some nomenclature is needed to understand the procedure and results. A run starts when the spinner starts rotating and ends when a bubble forms and a detection occurs or after sixty seconds have passed, whichever comes first. This sixty-second cutoff is needed to ensure the spinner does not rotate indefinitely and was chosen to keep the detector temperature down. The run time starts when the spinner has reached a high enough RPS that the sensitive volume has achieved the desired negative pressure, as calculated by the software, and ends when the run ends. A test consists of multiple runs. The number of runs is chosen to ensure a low error for the wait time which is a "real time" value given by the software. The wait time is calculated as the sum of all the run times divided by the number of detections. Detections occurring before the desired negative pressure is reached are automatically thrown out. Also, any time a run ends because there was no detection, sixty seconds are added to the sum of all the run times, but a detection is not added to the total number of detections. The first ten runs are

deleted for each test as the system has to degas itself before accurate data can be collected.[12] Last, for this research a batch of tests will be referred to as an experiment.

The formal experiments began with a background test to ensure that no adjustment would be needed once the rest of the tests were completed. Next, a gamma experiment was completed to ensure gamma insensitivity. This was done using a 7 μCi and a 100 μCi external ^{137}Cs source. Third, neutron experiments were completed to characterize the system. The experiments were completed using an external 0.54 mCi ^{252}Cf source. Four separate neutron experiments were completed:

1. First, a repeatability experiment was completed to ensure the repeatability of the results using the developed method. The experiment entailed performing nine tests, ensuring the starting conditions and method for each test were identical, and comparing the wait times, average wait time, and standard deviation at the end.
2. Next, a temperature/leakage experiment was completed to investigate the effects of temperature and fluid leakage on the run times. The experiment was completed by running the system for two hours and analyzing the trend of the run times.
3. Third, a distance experiment was completed to compare the results of varying the distance between the sensitive volume and the neutron source to results calculated using the inverse square law to further characterize the system. The experiment entailed performing three tests, increasing the distance of the neutron source from the sensitive volume between each test. The wait times

from this experiment were inverted to become an activity and compared with the inverse square law calculated values.

4. The fourth neutron experiment varied the pressure in the spinner between each test to characterize the spectroscopy abilities of the CTMFD.

The last formal experiment was completed using depleted uranium. The uranium was dissolved in acetone and placed directly in the spinner. Several tests were performed using different concentrations of uranium to test the ability of the CTMFD to determine the content of the fluid. An outline of the formal tests can be found in the "Testing Procedure" section to follow.

The above tests were chosen to best characterize the system. The gamma experiment was completed to confirm the claim of gamma insensitivity found in the literature.[10] The neutron experiments were performed to test the temperature dependence of the system, test the repeatability of results, and characterize the spectroscopy capabilities of the system. The alpha experiment was performed to test the quantification abilities of the system.

3.1 Experimental Equipment

The CTMFD used for these experiments was acquired from Sagamore-Adams Laboratories, LLC. The design of the system can be seen in Fig.4 with the main components numbered. The main components of the CTMFD are the following:

1. Rotary tool used to spin the system.

2. Speed sensors used to determine the current RPS. This is achieved by applying electrical tape and reflective tape to the spinner top of the spinner as seen in Fig.5. The speed sensor consists of an infrared LED and a light sensor placed next to each other at the top of the spinner. The LED constantly emits infrared light, but the light sensor only detects light when it is reflected off of the reflective tape. The amount of time between reflections is used to determine the RPS.
3. Glass Spinner which is the main component of the CTMFD.
4. Bubble sensors used to detect when a bubble has formed in the sensitive volume. This is achieved using an LED and light sensor placed on opposite sides of the sensitive volume. When a bubble forms in the sensitive volume the quality of light the sensor receives from the LED will change and the system will register a count.
5. Plastic enclosure used to protect the operator in case of catastrophic failure.
6. Base of the CTMFD which has adjustable rubber pads to keep the system stable.
7. Electronics box, seen in Fig. 6, which contains all of the electronics needed to run the system.
8. Pure acetone was used as the working fluid as suggested in the system manual.[12]

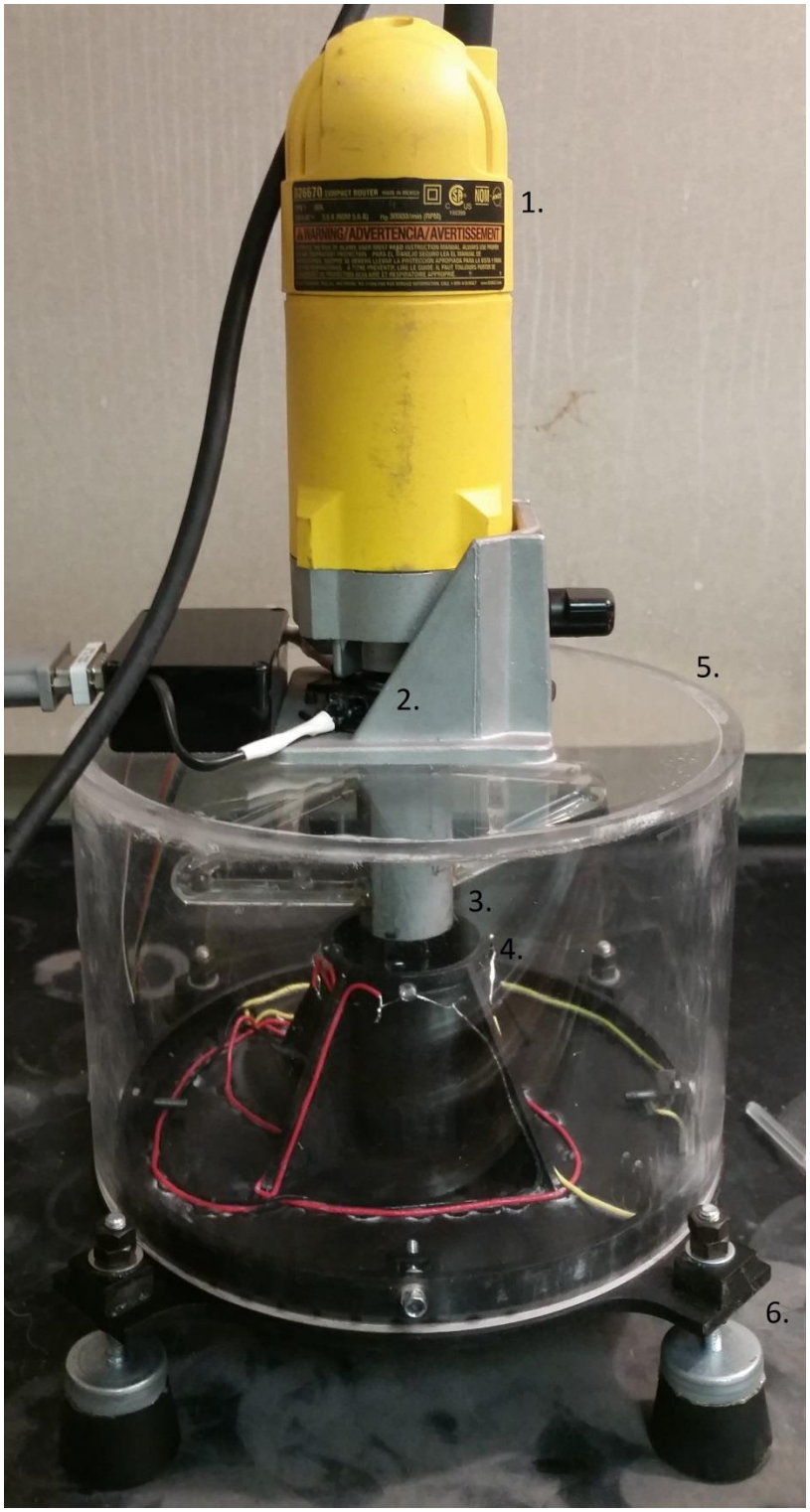

Fig. 4. The CTMFD System Used for Experimentation

Fig. 5. CTMFD Spinner with Tape Attached

Fig. 6. Electronics Box Containing Electronics Needed to Run the CTMFD

3.2 Testing Procedure

A procedure for running the desired experiments and methods for completing each test were developed at the beginning of experimentation.[12] The procedure developed for completing the formal tests was:

1. Set up CTMFD in an appropriate location with the proper safety compliance.
    a. Install provided software on a computer and attach computer to the electronics box.
    b. Ensure system is setup in radiation control area and with appropriate precautions for flammable liquids in case of leakage.
    c. Ensure proper procedures are followed for the handling and disposal of radioactive material.
2. Run background tests at -8 bar.
3. Run neutron experiments using an external 0.54 mCi ^{252}Cf source.
    a. Analyze the repeatability of system results by running a repeatability test which includes nine tests run at a pressure of -6.5 bar with the neutron source 28 cm away from the sensitive volume. A cool down time of one hour was used between each test and the spinner was emptied and refiled to ensure the same starting conditions. The source was removed and system shutdown when the tests could not be completed in one day. The next day, the system was restarted and the source was replaced on a mark,

made before removal, to ensure the neutron source was in the same position.

b. Analyze the effects of fluid leakage and temperature change by running a two-hour long test and analyzing the results. The neutron source was placed 28 cm from the sensitive volume. A second two-hour long test was run after the data from the first test was analyzed and changes were made to the detector.

c. Run three tests varying the neutron source distance to test inverse square law. A cool down time of one hour was used between each test and the spinner was emptied and refiled to ensure the same starting conditions. The system was run at -8 bar, and the distances used were 28, 43, and 58 cm.

d. Run eight tests at negative pressures of 4.5, 5.0, 5.5, 6.0, 6.5, 7.0, 8.0, and 9.0 bar to characterize the detector response. The neutron source was placed 28 cm from the sensitive volume. A cool down time of one hour was used between each test and the spinner was emptied and refiled to ensure the same starting conditions. The source was removed and system shutdown when the tests could not be completed in one day. The next day, the system was restarted and the source was replaced on a mark, made before removal, to ensure the neutron source was in the same position

4. Run detector with external 7 and 100 mCi ^{137}Cs gamma-ray source to determine gamma insensitivity. The gamma-ray source was placed 28 cm from the sensitive volume. A cool down time of one hour was used between each test and the spinner was emptied and refiled to ensure the same starting conditions.

5. Run alpha-particle experiments using depleted uranium dissolved in the active fluid.

    a. Prepare alpha solutions by dissolving UO_2 in acetone. Create several different dilutions to test the quantification abilities of the CTMFD.

    b. Test each dilution at a pressure of -9 bar, starting with the lowest concentration, and analyze results. The concentrations of uranium in the solutions used were 81.5, 59.0, 36.4, 28.5, and 20.6 ppm. The spinner was emptied and rinsed five times between each test to ensure no contamination.

The method developed was the following:

1. Turn off the rotary tool, turn off the electronics box, and unplug the black box from the wall. All power to the rotary tool must be turned off to thoroughly ensure the detector does not spin-up while the spinner is being prepared.
2. Unscrew and detach the plastic enclosure from the base ensuring the sensor cables are detached first as seen in Fig. 7.
3. Detach the glass spinner from the rotary tool by pressing the release button on the side of the rotary tool and unscrewing the spinner.

4. Unscrew and remove the top of the spinner and remove the rubber stopper.
5. Place the spinner in a cradle and fill to approximately the black lines with acetone as seen in Fig.5.
6. Replace the rubber stopper, the top of the spinner, and the screws.
7. Measure the fluid level distance in cm.
8. Place a layer of electrical tape along the top of the spinner as shown in Fig. 5.
9. Place a piece of reflective tape on the electrical tape as shown in Fig. 5.
10. Reattach the spinner by pressing the black release button on the side of the rotary tool and screwing the spinner in.
11. Attach the detector housing to the base ensuring the screws are replaced and the sensor wires are reattached as shown in Fig. 7.
12. Turn on the rotary tool, plug the electrical box into the wall, and turn on the electrical box.
13. Run the software as shown in Fig. 8 with each step lettered.
    a. Ensure the correct USB port is selected
    b. Start the program
    c. Ensure the bubble signal is working. If it is not, switch to the second sensor using the “Bubble Sensor” button.
    d. Enter the fluid level distance in cm.
    e. Enter the desired negative pressure in positive bar units. For example, to run a test at negative eight bar, a value of eight should be entered.
    f. Press the “Automation ON” button to start the test.

g. Ensure the RPS sensor is working

h. Ensure the threshold is appropriately set so that the bubble signal has a value above the threshold and a bubble formation causes the bubble signal value to drop below the threshold.

i. Press the “Automation OFF” once the desired number of runs have been made (k.).

j. Stop the program and save the data.

14. Wait and allow the spinner to cool down.

15. Repeat steps one through four and empty the glass spinner.

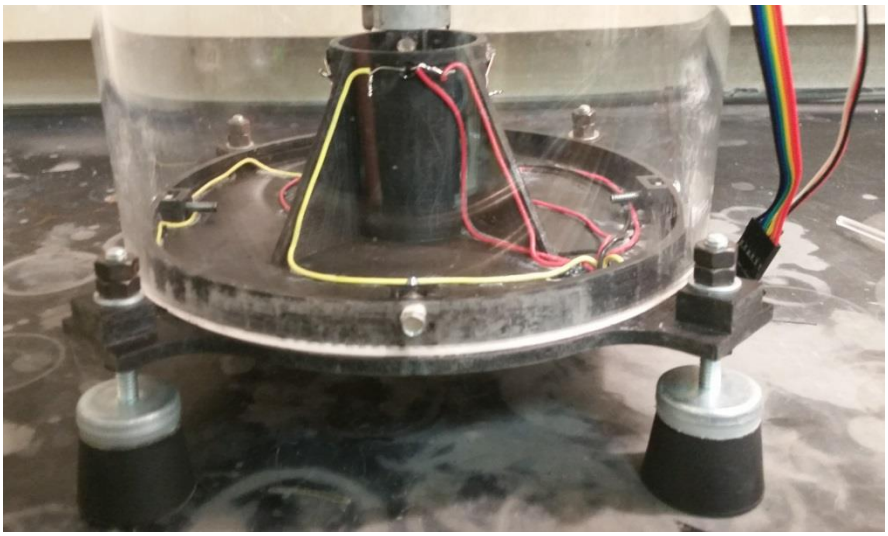

Fig. 7. Base of CTMFD with Sensor Wires

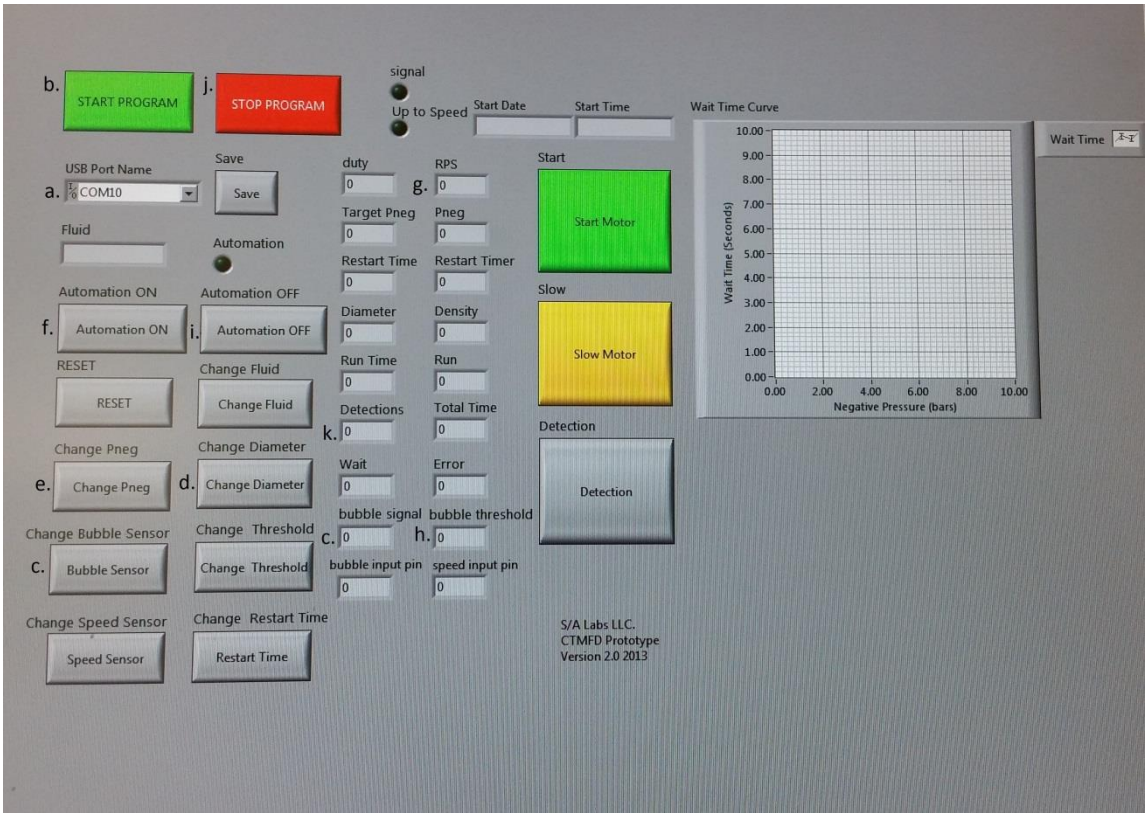

Fig. 8. Software Used to Control CTMFD

After several tests have been completed, the data are analyzed. In the tests, both the run time and wait time will be used for different purposes. After each test, the program saves an excel file that contains the following data for each run: run number, RPS at time of detection, pressure at time of detection, run time, number of detections so far, sum of run times so far, wait time so far and the wait time error. The current wait time and wait time error are shown on the program interface to determine when to end each test.

4. EXPERIMENTAL RESULTS

4.1 Background and Gamma Experiment

A background test was conducted first to ensure that background effects were statistically insignificant due to the very low background levels of radiation that could cause a bubble to form in the detector (essentially due only to neutron and cosmic ray background). This test was completed using a pressure of negative eight bar and a cool down time of fifteen seconds. For this test, fifty runs were performed over one hour. At the end of the test there were zero counts.

Next, a gamma-ray experiment was performed. Three tests were performed with two source strengths and two negative pressures. The first two tests were completed using a pressure of negative seven bar and the final test was completed using negative eight bar. A cool down time of fifteen seconds was used for all three tests. The gamma-ray source was placed 28 cm from the sensitive volume. Approximately forty runs were performed for each test. The first test used a ^{137}Cs source with listed activity of seven microcurie and the second two tests used a ^{137}Cs source with listed activity of one hundred microcurie. All three tests returned zero counts. This result was expected as gamma rays of this energy would have a low probability of interaction in the small sensitive volume of the detector and even if they did interact they would be unlikely to deposit energy in a high enough density to form a bubble.

4.2 Neutron Experiments

All neutron experiments were performed using a 0.54 mCi ^{252}Cf source. The distance and pressure were varied depending on the test. The temperature was taken inside the detector housing at the beginning and end of each test to ensure enough time had passed for the system to reach room temperature again. Initial tests were performed using the neutron source to ensure a familiarity with the system.

4.2.1 Repeatability Experiment

A repeatability experiment was used to estimate the repeatability of results from the testing method. The experiment was completed using a pressure of negative 6.5 bar and a cool down time of fifteen seconds. The neutron source was placed 28 cm from the sensitive volume. Approximately seventy-five runs were performed for each test. The wait times for each run are plotted in Fig. 9.

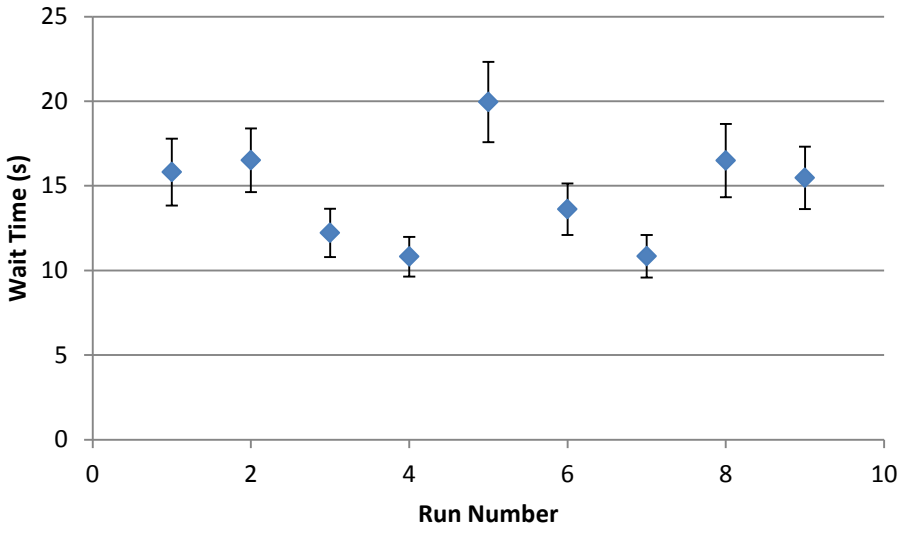

Fig. 9. Repeatability Test Showing the Precision of the Testing Method

The repeatability tests had an average wait time of 14.63 seconds with a standard deviation of 2.85 seconds. There was no discernable pattern in the results and they appeared to be randomly distributed about the mean. The testing method was deemed acceptable as all of the tests points were within two standard deviations of the average. This provided confidence that the between run variations were due to stochastic errors in the counts and not due to systematic variations or drifting effects. This testing method was used for the rests of the experiments but was slightly modified after the leakage tests.

4.2.2 Temperature and Leakage Experiment

During the repeatability experiment, it was noted that the fluid level distance lowered during experimentation and that the temperature of the system rose. It was

decided a longer test was needed to determine how the system was affected by temperature change and leakage. A single test was performed that ran for four hours; approximately four times longer than the repeatability tests. The test was completed using a pressure of negative 6.5 bar and a cool down time of fifteen seconds. The neutron source was placed 28 cm from the sensitive volume. The results of the test can be seen in Fig. 10.

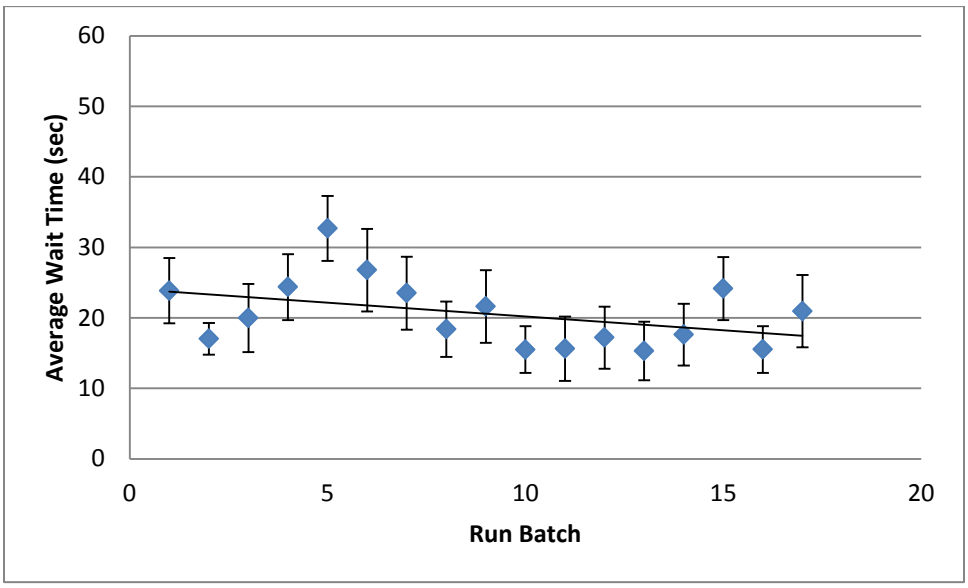

Fig. 10. Test Showing the Effects of Temperature and Leakage

The run times for the test were averaged into batches of twelve to provide better statistics. The slope of a linear fit line was then used to determine the effects of leakage and temperature. For the test, the slope was -0.39 with an R-squared value of 0.17. This shows that there was a minimal effect on the run times for the system.

A couple of changes were made to the system to mitigate leakage and minimize temperature rises because later tests would need to be performed at a higher negative pressure. First, the time between runs was increased from fifteen seconds to sixty seconds to ensure the system could cool down between runs. Second, a cap was placed above the rubber stopper and the rubber stopper was covered with vacuum grease to prevent it from rising out of the spinner and letting fluid leak.

A second test was taken after the changes had been implemented. The parameters were the same as the first leakage test aside from the aforementioned changes. The results of the test can be seen in Fig. 11.

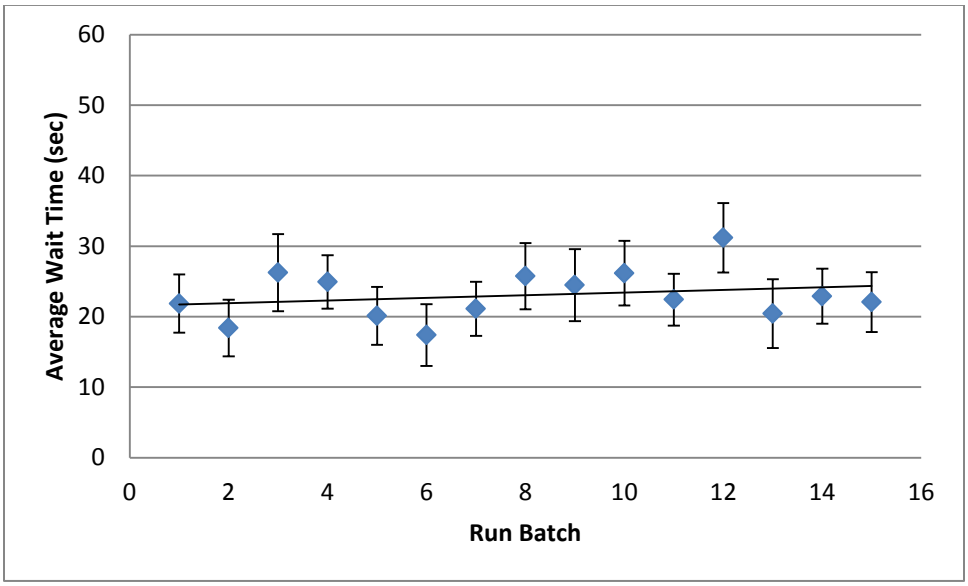

Fig. 11. Test Showing the Effects of Temperature and Leakage After Changes

For the second test, the slope was 0.19 with an R-squared of 0.06. Due to the slopes of both trend lines being comparable, it was hypothesized that the slope of the

trend lines is most likely do to inherent uncertainties of the system rather than directly contributed by leakage or temperature rise.

4.2.3 Distance Experiment

The next experiment varied the distance of the neutron source from the sensitive volume; the results can be seen in Fig. 12. The purpose of this experiment was to help characterize the system by comparing the results to the inverse square law. The experiment was completed using a pressure of negative eight bar and a cool down time of sixty seconds. A higher negative pressure was used to increase the sensitivity of the system. Approximately sixty runs were performed for each test.

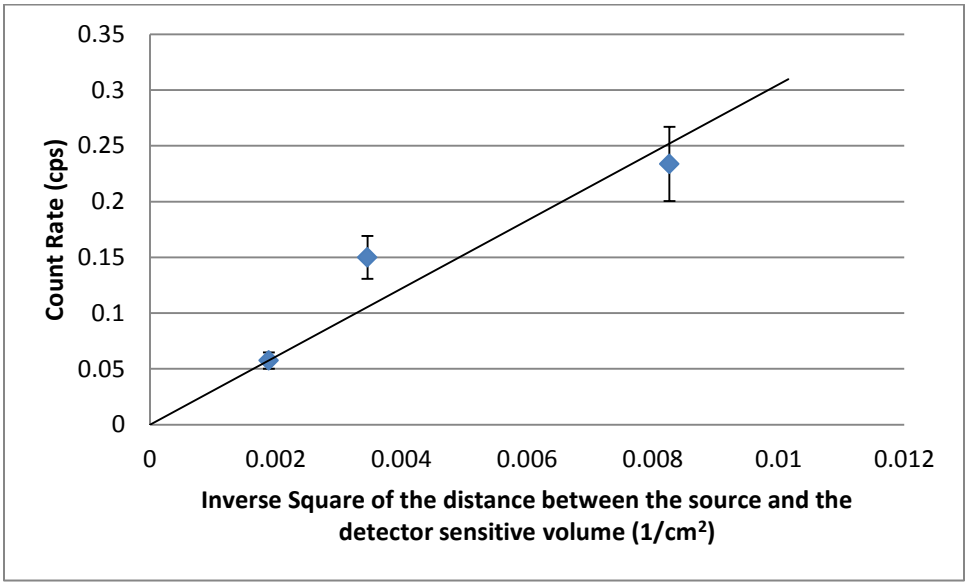

Fig. 12. Demonstration of Inverse Square Law

The results show that the system fairly well follows the inverse square law which is excellent. However, a high negative pressure was needed to obtain usable data for the farthest distance. At this negative pressure, the CTMFD may produce unwanted detections from other radioactive sources, if they are near.

4.2.4 Pressure Experiment

Once a solid understanding of the system was attained, an experiment was performed with a different pressure used for each test. The pressure experiment was performed to characterize the spectroscopy capabilities of the CTMFD. The experiment was completed using a cool down time of sixty seconds. The neutron source was placed 28 cm from the sensitive volume. Approximately seventy five runs were performed for each test. Eight tests were done with negative pressures of 4.5, 5.0, 5.5, 6.0, 6.5, 7.0, 8.0, and 9.0 bar. The wait time for each test was plotted on a log-log plot in Fig. 13 and the run times for each test were plotted as histograms in Fig. 14.

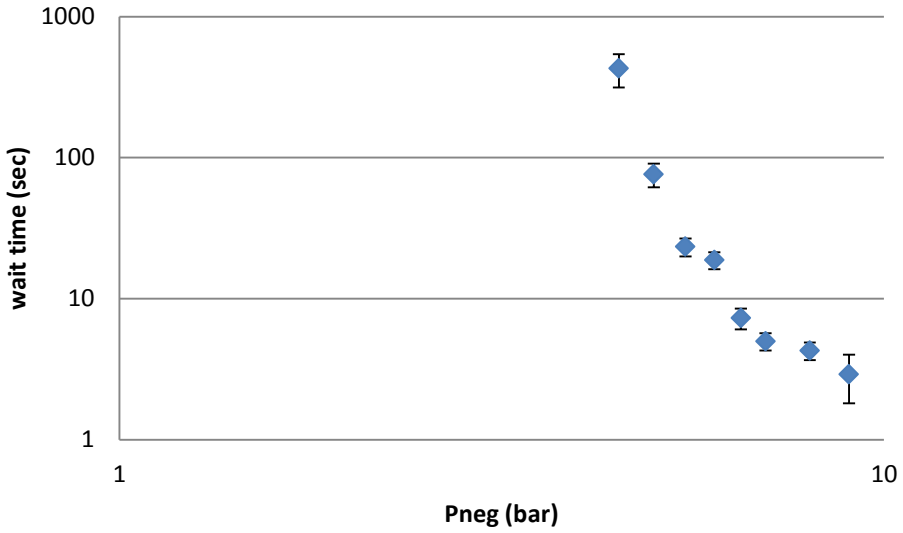

Fig. 13. Series of Tests Showing the Effect of Increasing the Negative Pressure

As seen in Fig. 13, as the negative pressure was increased, the wait time decreased. This phenomenon was expected as at higher negative pressures the energy required to form a bubble is lowered and the lower energy neutrons from the ^{252}Cf spectrum can cause bubbles. This can be more clearly seen when comparing Fig. 14, histograms of the run times for each test, and Fig. 15, the ^{252}Cf neutron energy spectrum.[13]

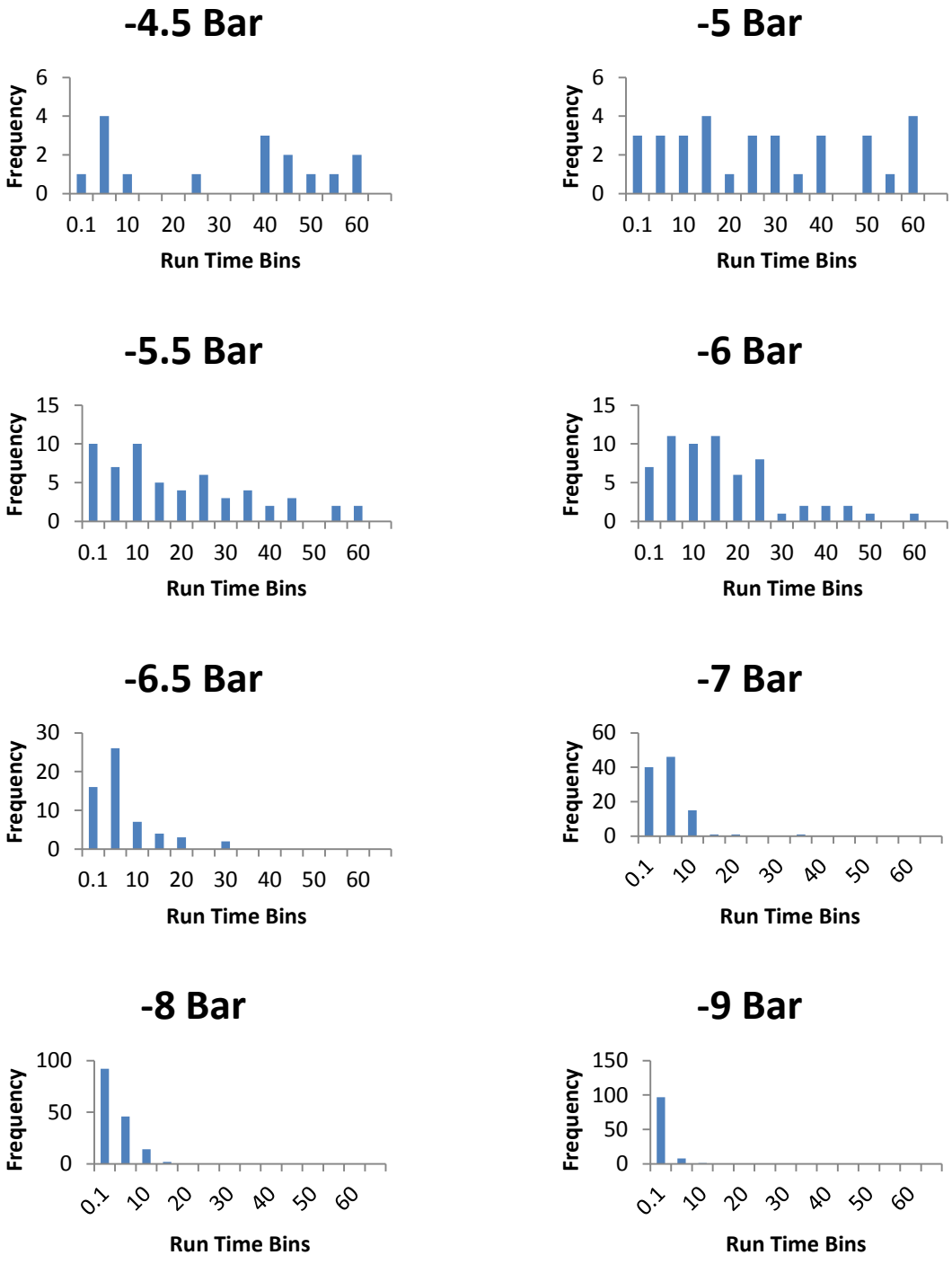

Fig. 14. Histograms of the Tests Showing the Effect of Increasing Negative Pressure

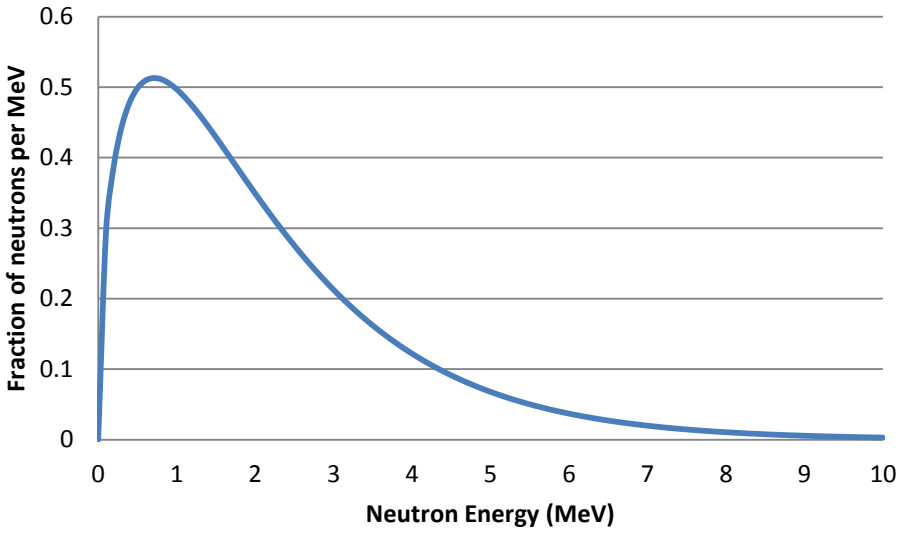

Fig. 15. Maxwell Distribution of ^{252}Ca Neutron Spectrum [13]

When looking at the histograms a pattern emerges. It can be seen that as the negative pressure increases the run time decreases. This phenomenon is due to the lower energy needed to form bubbles in the sensitive area. Increasing the negative pressure allows more neutrons to be detected in the system as lower energy is needed to create a bubble. For ^{252}Cf, the CTMFD would be unable to determine the shape of the spectrum at very low energies because the one and two MeV neutrons would desensitize the system. The effects of higher energy neutrons on the system will decrease the effectiveness of the CTMFD as a spectroscopy instrument. This effect can be more easily seen below.

Fig. 16 shows a sample spectrum of two speculative neutron emitters. As the CTMFD reached the negative pressure required to form bubbles from the five MeV neutrons, the wait time for the system would be similar to the "-9 bar" chart. When the

negative pressure is increased further, the five MeV neutrons would continue to have very short wait times, desensitizing the CTMFD to the lower energy neutrons. The effects of five MeV neutrons on the system would cause the spectrum from the CTMFD to look similar to Fig. 17, where the second neutron peak is completely missing due to the large effect of the five MeV neutrons. These effects could cause the CTMFD to be unusable for spectroscopy if multiple energy sources are present.

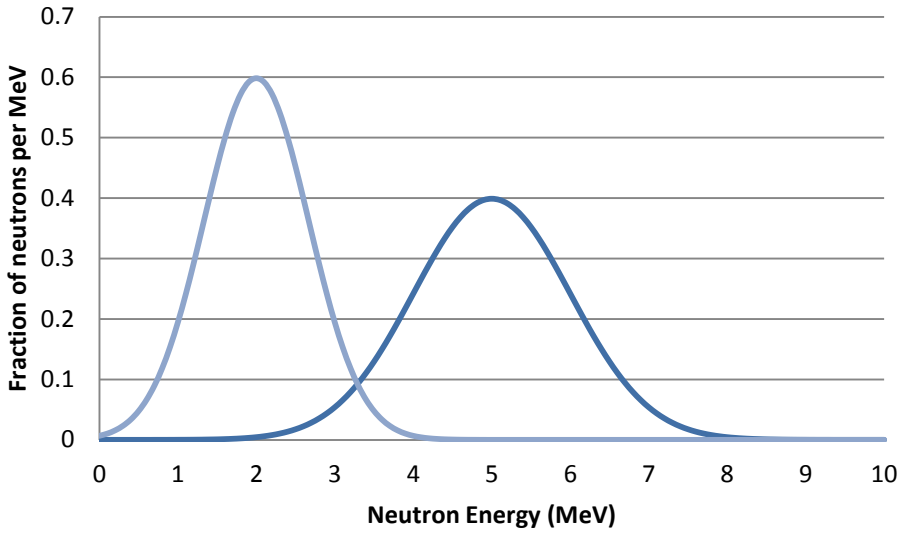

Fig. 16. Speculative Maxwell Distributions

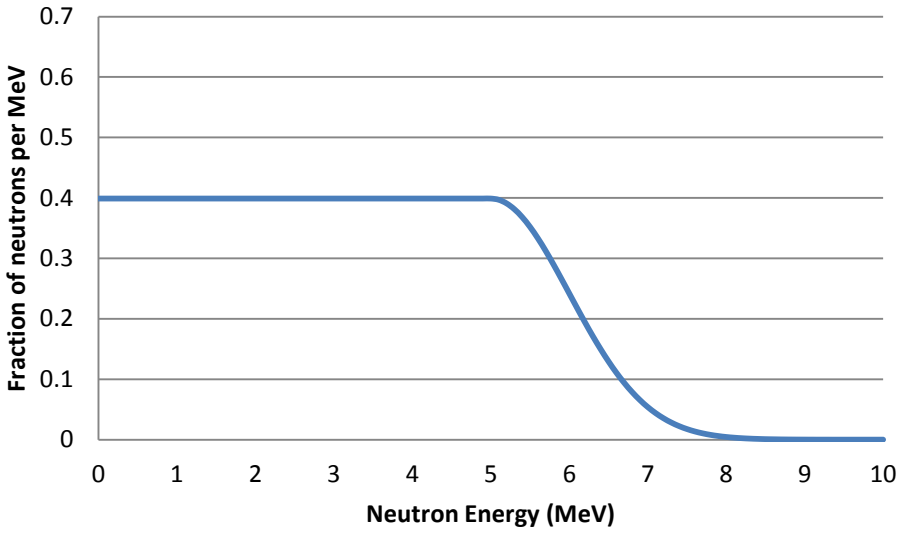

Fig. 17. Sample Spectrum Due to Desensitization From a CTMFD

4.3 Alpha Experiment

The last experiment performed was an alpha experiment. Alpha detection is one area where the CTMFD could have advantages over currently available detectors. As has been stated, the CTMFD works by placing a working fluid in the glass spinner. For all of the previous tests, this liquid was pure acetone. However, the working fluid can contain other things such as an alpha or beta emitter. For alpha tests, the radioactive source to be measured could be placed directly in the CTMFD if it was a liquid, or it could be dissolved in the acetone if it was a solid.

For this experiment, a small sample of depleted UO_2 was dissolved in the acetone. The uranium-acetone solution (U-A solution) was diluted to create several different concentrations to test. These concentrations were placed directly in the CTMFD which was run following the same method as described in this thesis.

The first test performed used the highest activity U-A solution which was 68.3 Bq/ml. The test was performed to determine the required negative pressure for the rest of the alpha tests. It was originally estimated that a low negative pressure could be used because ^{238}U has an alpha decay of 4.27 MeV and it was believed that all of this energy would be deposited in the sensitive volume.[7] However, after testing, it was found that a negative pressure of at least nine bar was needed. It was determined, through a literature search, that it was not the alpha particle energy that was responsible for the bubble formation, but that it was the recoil nucleus energy.[11] The alpha particle simply did not deposit energy in a high enough density to cause bubble formation. Thus the energy deposition per decay was much lower than initially expected and this resulted in higher negative pressures needed.

The rest of the alpha tests were performed at a negative pressure of nine bar. However, at this pressure, the rotational speed of the spinner caused the CTMFD to vibrate significantly. Lead bricks were placed around the base of the CTMFD to keep it from vibrating across the lab table. Also, the vibration was so intense that the bubble formation sensor stopped working and all bubble formations had to be recorded manually. It was desired that the tests be run at a higher negative pressure, but because of the vibration, the pressure was kept as low as possible.

After the initial test was performed a more strict method was developed to help counter the effects of the high RPS. This method included a person being present at all times during the test too ensure all bubbles were counted and the RPS sensor continued to work, and one test was performed at the same time each day to ensure the temperature

profile was the same for each test. The results of these tests (measured count rates versus uranium concentration) can be seen in Fig. 18.

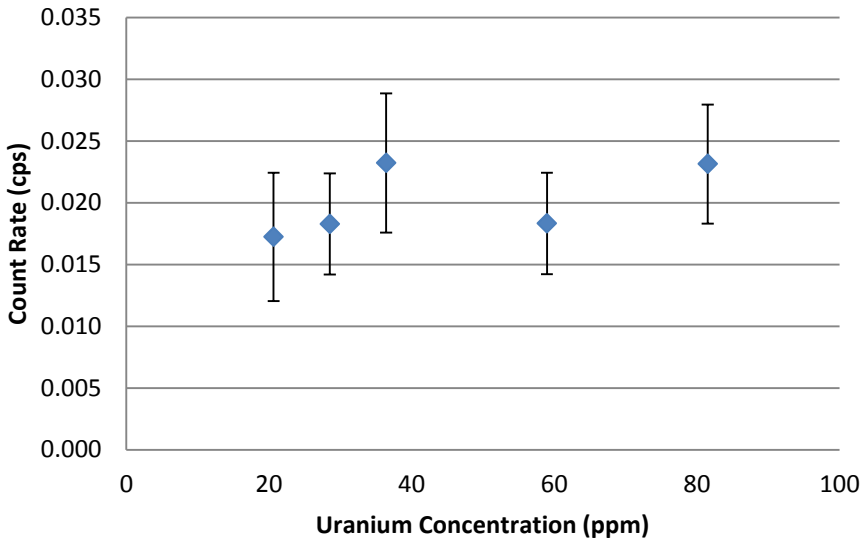

Fig. 18. Tests Showing the Alpha Detection Ability of the CTMFD

As can be seen, within error, the count rate did increase with the amount of uranium as expected. However, the error bars are very large because, even at a pressure of negative nine bar, most runs were ending at the sixty second cutoff instead of registering a count. It is also important to note that this data does not appear to extrapolate to a y-intercept of zero suggesting that the statistical errors in the data are far too large. The alpha experiment proves that in theory the CTMFD could be used as an alpha detector, but more work is needed to refine the design of the system for such an application.

4.4 Sources of Error

During experimentation several sources of error were discovered. The first two sources of error were from temperature rise during testing and leakage of acetone during testing. The temperature/leakage experiment was conducted to study the error from these sources and to try and diminish it. However, because the alpha experiment was run at a high RPS, the temperature of the system still rose and could have an impact on the alpha experiment results. The high RPS also caused the spinner to vibrate, which caused the detection sensor to fail leading to the need for user input to record detections. The speed sensor works similarly to the detection sensor leading to possible errors in the RPS which is used to calculate the pressure. Human error is another contributor and comes from two main sources. First, because the system is very sensitive to the fluid level distance any errors in the preparation of the spinner before every test could cause error in the results. Second, errors could have been made in the preparation of the uranium acetone solutions causing incorrect concentrations. Next, the inability of the system to count beyond sixty seconds could cause error in the detections. Last, bugs in the system software could cause errors. The results from the software gave some unexpected values including values of negative one for the negative pressure, detections that occurred at pressures much more negative than the desired negative pressure, and counts occurring past the sixty second cutoff.

5. CONCLUSION

Through experimentation it was determined that the CTMFD has great promise as a novel, real-time detector system. It was determined that the detector was gamma insensitive, at low activities for external gamma sources. Neutron experiments also showed that the detector system could be used as a neutron detector but it is unclear that it has significant advantages over conventional systems. Where the CTMFD shows real promise is with alpha detection. During these experiments, it was found that the system could detect small amount of uranium within seconds, but that further work is needed for the system to be used for safeguards in a reprocessing facility (where accurate quantification is vital).

One of the main interests of this work was for the application of the detector system to online measurement at the dissolver tank and product lines of a reprocessing facility. The current technology may have difficulties in this application but more study is needed. The experimentation showed that neutron detections occurred at much lower negative pressures than alpha detections making the system unusable for the dissolver tanks because of the large neutron field created by the actinides. A significant study would be needed to determine how a system could be designed to compensate for this neutron effect. However, the TMFD's ability to detect minute amounts of uranium in solution could make it an ideal detector for the product lines which mainly contain only uranium and plutonium.

Some future work is needed in order to fully characterize the CTMFD's safeguards capabilities. Beta experiments need to be performed in order to analyze the

CTMFD's beta detection capabilities. Also, higher intensity gamma, neutron, and alpha sources need to be tested in order to simulate the dissolver tank and product lines of a reprocessing facility. Lastly, gamma-ray and neutron sources need to be inserted in the working fluid, similar to the alpha sources, in order to test the ability of the recoil nuclei from the creation of the gamma-rays and neutrons to cause bubbles to form.

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